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Exciton formation and relaxation in GaAs epilayers

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Exciton formation and relaxation in GaAs bulk epilayers have been studied by means of time-resolved photoluminescence techniques. It is found that the time evolution of the free exciton luminescence, nonresonantly excited at low temperature and low intensity, is extremely slow, with a rise time of the order of 1 ns and a decay time of several ns. Simulations based on Monte Carlo solution of the set of coupled Boltzmann-like equations for free carriers and excitons show a nice agreement with the experimental data, and suggest a dominant role played by acoustic phonons in the exciton relaxation. [S0163-1829(98)53044-5]

The energy relaxation of photogenerated electron-hole pairs plays a central role in modern semiconductor science. The time evolution of the carrier population created by band-to-band ultrashort laser pulses is determined by scattering processes with other elementary excitations; therefore, information on the fundamental interactions in semiconductor materials can be directly obtained from time-resolved optical measurements.[1] The very same scattering processes determine the response of high-speed devices and provide the basis for the design and development of most ultrafast electronic and optoelectronic applications. The enormous activity dedicated to this topic is therefore motivated not only by the interest in fundamental physical phenomena, but also by the importance of carrier energy relaxation mechanisms in applied semiconductor science.

Most of the work has been devoted so far to the hot carrier regime after high power excitation, where many-body effects and nonequilibrium and/or nonthermal carrier distributions play the major role.[2] Much less attention has been paid to the low excitation regime and, in particular, to the details of exciton formation from electron-hole pairs, in spite of the fundamental relevance of the excitonic features to the optical properties of direct-gap semiconductors. Almost all of the published experiments deal with exciton formation and relaxation in GaAs-based quantum wells (QW's);[3] little is known of the behavior of thick bulklike layers, which is important also as a reference for understanding nanostructures. The experimental data are analyzed within phenomenological rate equation models and a detailed comparison with microscopic calculations is still missing. The following qualitative picture has been shown to apply to the case of QW's: the photogenerated electron-hole pairs tend to form excitons with large in-plane momenta on a fast time scale ($<$20 ps). The subsequent thermalization and relaxation of the exciton ensemble is much slower (hundreds of ps) and is mainly determined by exciton-acoustic phonon and exciton-exciton interaction.[4] Only recently theoretical studies have started addressing the problem of exciton formation and dissociation in both bulk and GaAs-based QW structures on the basis of microscopic calculations, by solving coupled Boltzmann equations for electrons, holes, and excitons.[5–7] However, these papers concentrate on the energy relaxation processes in the initial 200 ps, that is, the time scale where exciton formation and relaxation occur in QW heterostructures.

In this paper we report on an experimental study of the exciton formation and relaxation in GaAs bulk epilayers by using continuous wave (cw) and time-resolved photoluminescence (PL) techniques, as a function of the excitation energy, temperature, and density. The data are compared with simulations based on a Monte Carlo solution of a coupled set of Boltzmann-like equations.[5] In particular, the theoretical analysis of Refs. 5–7 is extended over a 1-ns time window. We find that the time evolution of the free exciton PL signal, nonresonantly excited at low temperature and low intensity, is extremely slow, with a rise time of the order of 1 ns and a decay time of several ns. The long decay time demonstrates the good quality of the sample investigated while the 1-ns rise time is found to be a mere consequence of the relevance of acoustic-phonon interaction in the exciton formation and subsequent relaxation to radiative states. In fact, the comparison with the predictions of the microscopic theoretical analysis of Refs. 6 and 7 indeed shows a very nice agreement concerning the fundamental role played by acoustic phonons in the exciton relaxation on the ns time scale.

We have investigated two different molecular beam epitaxy grown GaAs epilayers, 1-μm thick and nominally undoped. We have used standard PL setups; cw PL and PL excitation (PLE) spectra have been performed by means of a tunable Ti:sapphire laser. The luminescence, dispersed by a

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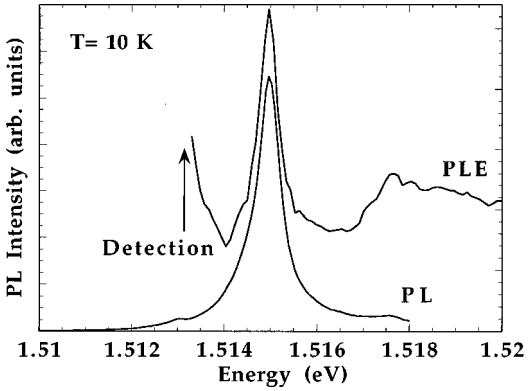

FIG. 1. Typical PL and PLE spectra at $T=10$ K and low excitation.

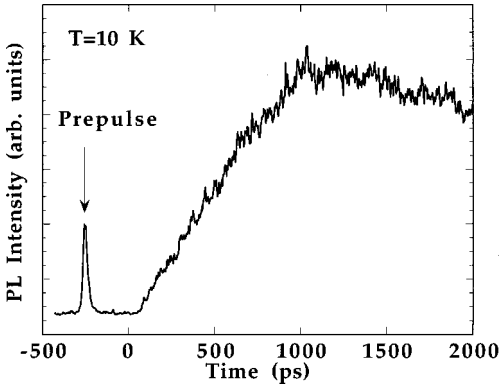

FIG. 2. Time evolution of the PL intensity at the free exciton recombination energy (1.515 eV) at $T=10$ K. The excitation is set at 1.560 eV; the excitation density is of the order of 10^{14} cm^{-3}.

60-cm double monochromator, has been detected by standard photon counting techniques. Time-resolved measurements have been obtained by using a synchronously pumped dye laser; the time duration of the pulses was 5 ps, with a repetition rate of 76 MHz. The laser beam was split in two parts: the first one (prepulse) was sent to an optical delay line for setting the time reference, the second one was used for the excitation. The luminescence and the prepulse were detected simultaneously by a streak camera in synchrosan operation with a time resolution of 20 ps and a time window of 1.5 ns. The samples were mounted on the cold finger of a closed-cycle cryostat in order to perform measurements down to 10 K.

The main ingredients of the simulations are the probabilities associated to the relevant scattering mechanisms, including phonon-mediated exciton binding and dissociation, as well as carrier-carrier, carrier-phonon, and exciton-phonon interactions. These are obtained from microscopic calculations as described in Refs. 5–7; no free parameter is therefore left in the simulations. Note that radiative and nonradiative recombination mechanisms are not taken into account; correlation between the photogenerated pairs and polaritonic effects are also neglected. The coupled kinetic equations of Boltzmann type are solved by means of an extended Monte Carlo method that treats the free-carrier–exciton reactions as many particle collisions.[5] This allows us to study in detail the time dependence of the free carrier and exciton populations in the incoherent regime, and use them to predict the time evolution of PL in off-resonant excitation conditions.

Typical cw PL and PLE spectra at $T=10$ K and low excitation are reported in Fig. 1. The comparison between the PL and PLE spectrum clearly shows that the PL signal is dominated by the free exciton recombination at 1.515 eV in both samples; the full width at half maximum of the excitonic resonances are of the order of 0.55 and 0.75 meV, respectively. These results point out the high quality of the samples investigated.

The temporal profile of the PL signal at the free exciton recombination energy, at $T=10$ K and with excitation at 1.56 eV, is shown in Fig. 2. In order to follow the slow PL kinetics of the GaAs bulk epilayers, two different time windows of the streak camera system, delayed by 1 ns, have been separately acquired. Note that the absolute value of the delay time is meaningless; we have chosen to set $t=0$ at the rising edge of the PL. However the presence of the prepulse, that with our setup occurs at $t=-300$ ps, fixes a common origin of the delay time for the whole set of measurements allowing an accurate comparison between different decay curves. The time evolution of the excitonic emission is extremely slow; we find a rise time of the order of 1 ns in both samples and we estimate the decay time to be around 4 and 5 ns, respectively. The values observed for the decay times are in good agreement with previous findings[8] in good-quality GaAs epilayers, which have been interpreted in terms of radiative decay of exciton polaritons through coupling to longitudinal-acoustic phonons.[9] We therefore exclude a major role, if any at all, of the nonradiative recombination channels in the exciton kinetics of the samples investigated. As far as the rise times are concerned, we are not aware of previous reports on the rise time of the excitonic PL signal in GaAs bulk samples. Such a long rise time, extending into the ns time scale, is quite unexpected if compared with the few hundred ps usually observed in GaAs-based quantum wells.

The temperature dependence of the temporal profile of the PL signal at the peak of the excitonic band is reported in Fig. 3(a). The rise time dramatically decreases when the temperature is raised, and for $T=50$ K it is already as short as 70 ps. For an easier comparison, the theoretical predictions, based on corresponding Monte Carlo simulations for electrons, holes, and excitons are reported in Fig. 3(b). The agreement between experiment and theory is indeed very good.

From the analysis of the simulations, which allow us to follow the time evolution of both the free-carrier and the exciton populations, it appears that the extremely long rise time is not due to the exciton formation, but instead to the slow energy relaxation of the hot excitons via acoustic phonons. The calculations indeed show that the electron-hole pairs, photogenerated within a vertical transition following photon absorption, rapidly decay into excitonic states. As a matter of fact, the calculated total exciton density (i.e., the distribution function integrated over the whole band) is very different from the PL showing a fast rise (<50 ps) to a nearly constant value. However, exciton formation with large center-of-mass momentum (hot excitons) is found to be much more favored with respect to the direct formation at the bottom of the excitonic band. The initial exciton distribution is therefore nonthermal, with a predominance of hot excitons that cannot emit a photon due to momentum conservation. The PL signal, which arises only from $k\sim 0$ exci-

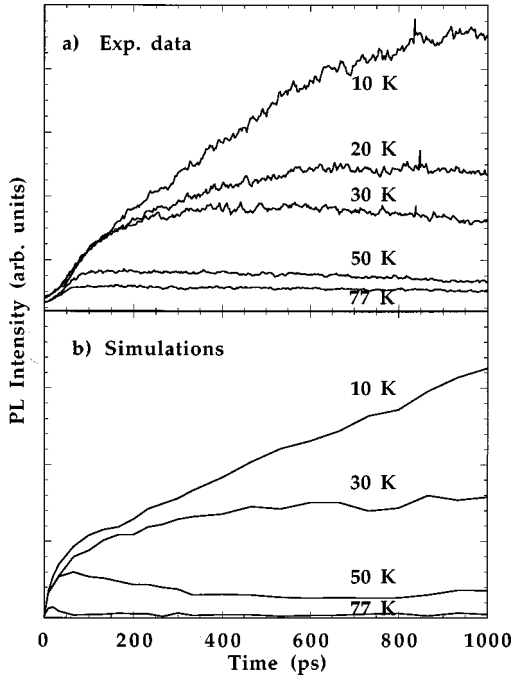

FIG. 3. Temperature dependence of the temporal profile of the PL signal at the peak of the excitonic band. (a) Experimental data. (b) Theoretical simulations using the model in Ref. 5. The excitation density is $\sim 10^{14}$ cm^{-3} in all cases.

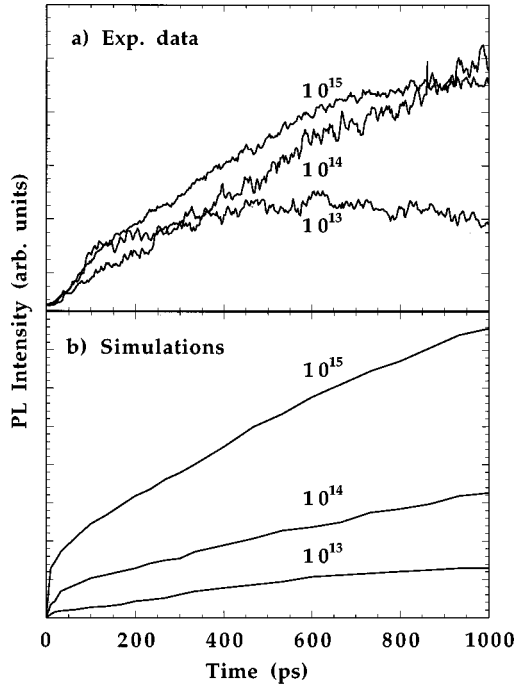

FIG. 4. Excitation density dependence of the temporal profile of the PL signal at the peak of the excitonic band, at $T=10$ K. (a) Experimental data. (b) Theoretical simulations using the model in Ref. 5. The excitation densities are given in units of cm^{-3}.

tons, rises therefore with the relaxation time of the hot excitons toward the bottom of the excitonic band. The simulation shows that this relaxation process is dominated by exciton-acoustic phonon interaction and it is therefore quite slow.

A comparison between experimental results and theoretical predictions for excitation densities varying over two orders of magnitude is reported in Fig. 4; the profiles are scaled by the excitation power for ease of comparison. The experimental excitation density has been estimated from the measured excitation power by using the values of the absorption and reflection coefficients of GaAs bulk reported in the literature. The agreement between theory and experiment is, in this case, only qualitative. In fact, while for each excitation density the experimental and theoretical PL time evolutions occur on the ns scale with similar rise times, the experimental data do not exhibit the superlinear increase of the PL intensity predicted by the theory, which is essentially due to bimolecular exciton formation rate. However, a quasilinear dependence of the excitonic PL signal is indeed expected in good quality samples where the internal radiative efficiency is of the order of 1. We believe that this discrepancy is mainly related to the fact that the present theoretical analysis completely neglects the recombination processes; moreover, it is obvious that the experimental situation never does correspond to the uniform excitation density across the laser spot assumed in the theoretical simulation.

The dependence of the PL rise time on the excitation energy is reported in Fig. 5(a) for excitation energies corresponding to either near-resonance or off-resonance conditions for the exciton formation via LO-phonon interaction. The striking point of the experimental results is that the resonant conditions with the LO phonon does not modify in a dramatic way the time evolution of exciton PL, as naively expected. The energy relaxation dynamics is in all cases dominated by the slow interaction with the acoustic phonons and the resonance conditions with the LO phonons is predicted to affect the evolution of only a small fraction of the total exciton population during the first 50 ps. On the contrary, as shown in Fig. 5(b), the excitation near the band edge

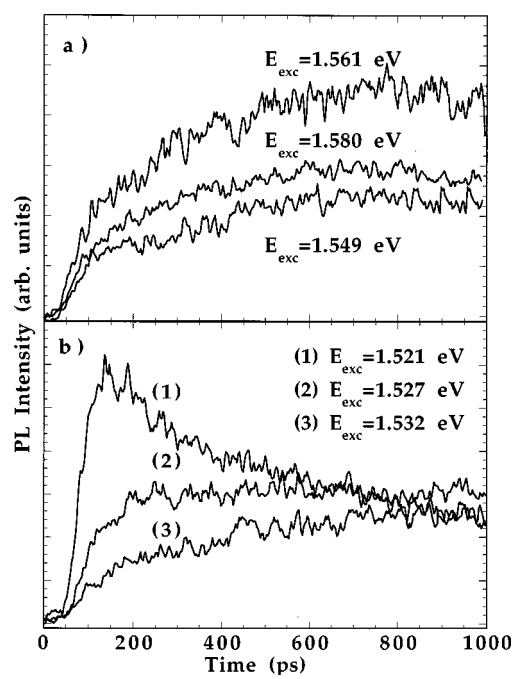

FIG. 5. Dependence of the PL rise time on the excitation energy E_{exc}, at $T=10$ K and an excitation density of $\sim 10^{13}$ cm^{-3}. (a) The excitation excess energy is scanned across the value of the LO phonon energy, (b) Near-resonant excitation.

strongly modifies the excitonic luminescence. We see that the rise time becomes faster and faster when the excitation energy approaches band-to-band absorption. According to previous results on the resonant fluorescence in GaAs bulk structures,[10] the fastening of the exciton luminescence kinetics for resonant excitation can be ascribed to the polariton nature of the bulk exciton. The microscopic model used for the theoretical simulation does not include, for the sake of simplicity, polariton effects as well as any correlation between the photogenerated pairs (geminated recombination) and therefore cannot reproduce the observed exciton dynamics for quasiresonant excitation conditions.

We would like also to add that time-resolved PL experiments are widely used as a powerful tool for investigating the carrier and exciton relaxation in semiconductor structures, owing to the fact that it is a simple, noninvasive, and linear optical technique. However, since the excitonic radiative emission only occurs at $k \sim 0$, the PL is unable to directly monitor the details of the excitonic distribution function, and this makes it quite difficult to extract direct quantitative information. We believe that the analysis we have presented, that is, a direct comparison between the experimental data and the theoretical predictions based on a microscopic approach, with no fitting parameter, represents a step further for a more detailed comprehension of the energy relaxation in semiconductor structures.

In conclusion, the time evolution of the excitonic emission from good-quality GaAs epilayers has been investigated as a function of temperature, excitation density, and excitation energy. We have unexpectedly found, at low temperatures, extremely long PL rise times up to 1–2 ns, well beyond the few hundred ps usually observed in GaAs-based quantum well heterostructures. The comparison with theoretical predictions, based on the Monte Carlo solution of a set of coupled Boltzmann equations for electrons, holes, and excitons, unambiguously allows us to relate the long PL rise times to the exciton relaxation to radiative states via acoustic-phonon emission. Indeed, a very good agreement between experiments and theoretical predictions is obtained at low temperatures: moreover, the dependence of the PL rise times on temperature is very well reproduced by the theoretical simulations.

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